Intermediate long-lived nuclear waste management: an integrated approach to assess the long-term behaviour of cement-based materials in the context of deep disposal

C. GALLÉ*

***** French Atomic Energy Commission (CEA, Saclay)**
Nuclear Energy Division / Department of Physico-Chemistry

Summary

- ***Introduction and general context***

- ***Overall strategy for concrete long-term behaviour (L-TB) studies***

- ***L-TB in unsaturated environment (interim storage)***

- ***L-TB in saturated environment (deep disposal)***

- ***Conclusion***

Contributors (LECBA Laboratory): H. Peycelon, P. Le Bescop, S. Bejaoui, V. L'Hostis, B. Bary, P. Bouniol, C. Richet

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- **General context of research activities**
  - French nuclear ILL & HL wastes management policy ➔ ruled by the Dec. 30, 1991 French Parliament law = 3 main lines of research (1991-2006):
    - ① Partitioning & transmutation (CEA)
    - ② Deep geological repository (ANDRA)
    - ③ Waste conditioning & long-term interim storage (CEA)

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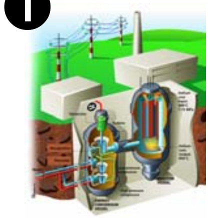

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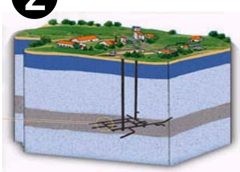

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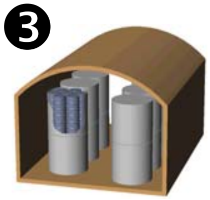
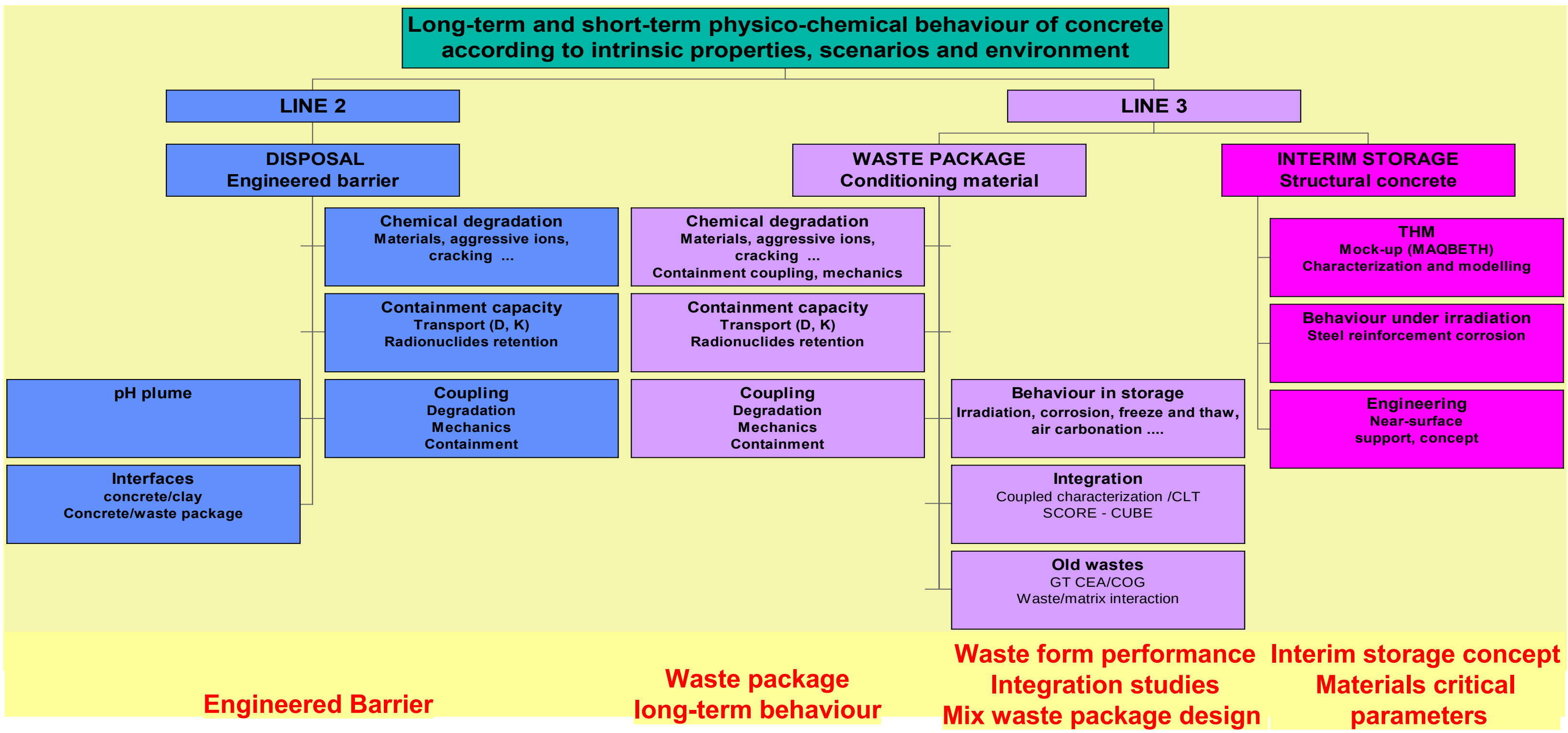
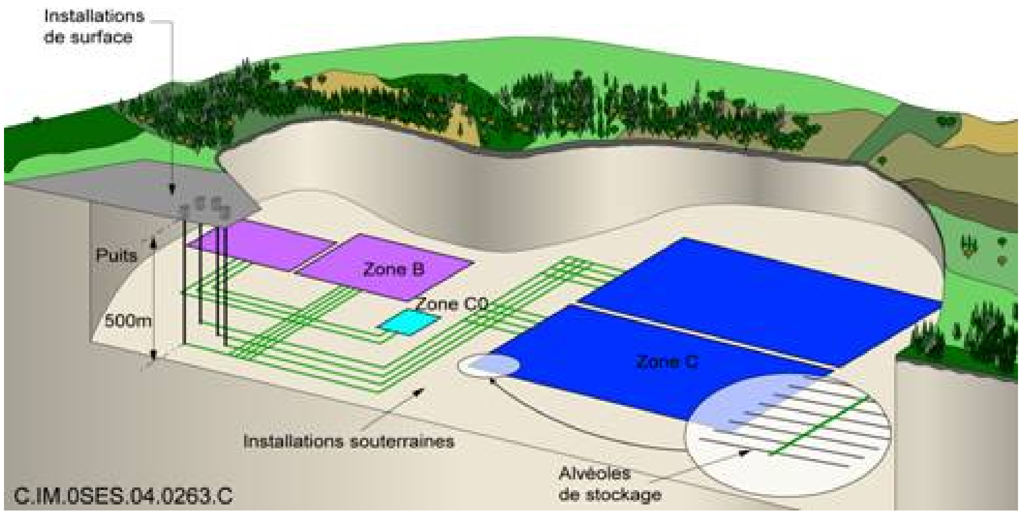
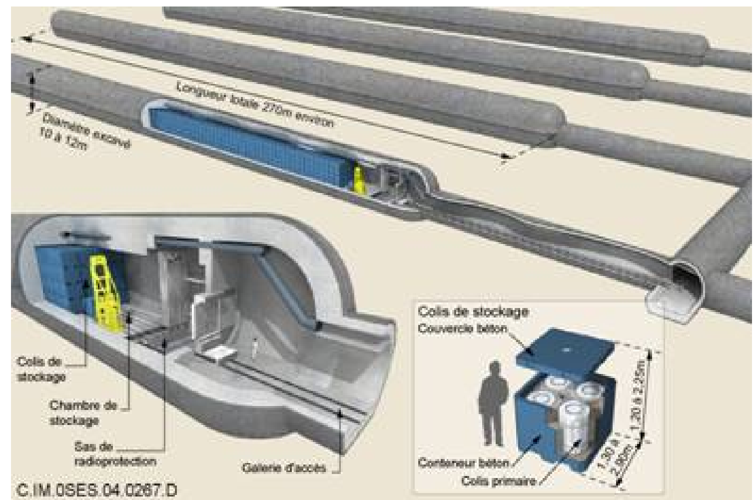




- **Second phase related to all radioactive wastes**
  - Since June 28, 2006 a new law determines the orientations of the research dedicated to waste management: main dead-line 2012 with a public debate
  - New lines of the law:
    - Spent fuel treatment: Partitioning and transmutation (leader CEA)
    - **Retreavable disposal in deep geological formation (leader ANDRA)**
    - Conditioning of wastes and temporary storage (leader ANDRA)





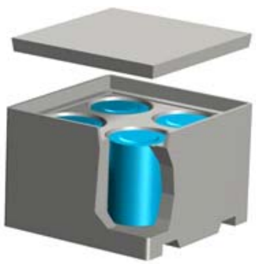
Disposal is now identified as the reference solution

Functional analyses (interim storage & disposal)

- **Safety assessment and performance analysis of the facilities**
  - To guarantee waste package (wp) confinement (no dispersion) and mechanical properties (wp recovery) during interim storage
  - To limit the radionuclides (RN) release during disposal phase

Storage design

- **Operational analysis (role of concrete + wp functions)**
  - Parameters to be evaluated at t_0 (wp integrity)
  - Parameters to be monitored during wp storage life-time
  - Most favourable storage conditions for wp
  - Recovery phase possible (300 years)?
  - Wp state at 300 years compatible with disposal phase entrance?



Mix storage/disposal design

- **Scientific questions**
  - Wp chemical degradation related to underground water leaching
  - Transport properties evolution during wp life-time
  - RN physico-chemical state and location with time
  - Cracking intensity and location predictability
  - Amount of H_2 gas generated by radiolysis
  - Corrosion rate and products in alkaline medium
  - Behaviour of organic matter in alkaline medium

Wp life-time environments / key phenomena & potential impacts

Evolution in unsaturated closed system & Evolution in unsaturated open system

- Alkali-aggregate reaction
- Delayed ettringite formation
- Waste / matrix interaction
- Radiolysis
- Mechanical stress

- Drying / resaturation
- Air carbonation
- Corrosion
- Freeze & thaw cycling
- Organic matter degradation

Evolution in saturated open system

Reactions leading to hydrolysis and leaching of hardened cement paste components (alteration by water, pH < 12, temperature 25 – 85°C)

- Increase of:
  - porosity
  - permeability
  - diffusivity

Reactions between aggressive fluids and hardened cement paste components leading to the formation of secondary phases

Non expansive products (carbonates)

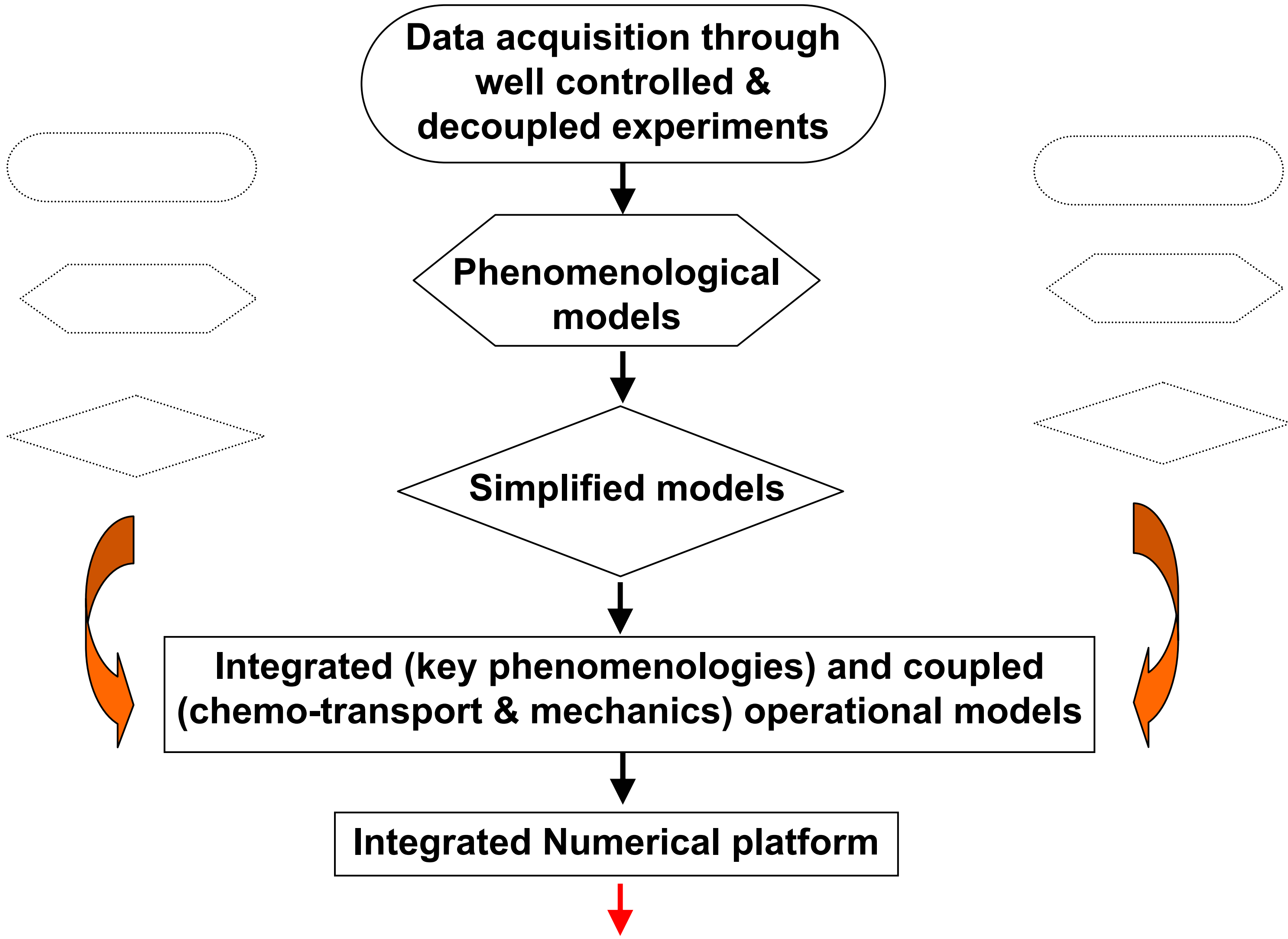
- Decrease of:
  - porosity
  - permeability
  - diffusivity

Expansive products (sulfates)

- Increase of:
  - internal stress
  - microcracking

Loss of Alkalinity	Loss of weight	Decrease of strength & stiffness	Cracking & spalling	expansion deformation

Basic operational modelling strategy



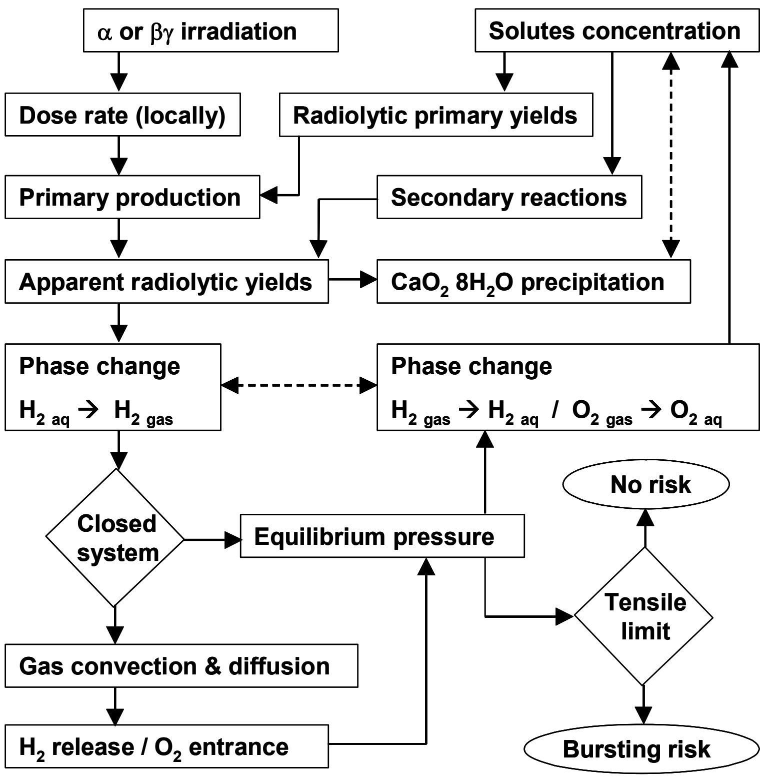
Waste package physical & mechanical states description + RN migration

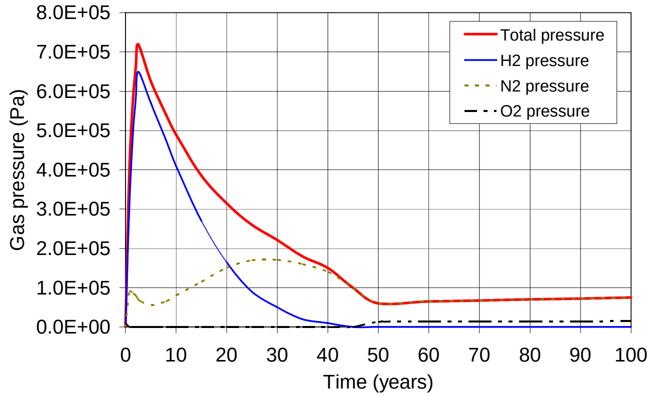
- **Key identified topics for the wp long-term storage evolution**
  - Radiolysis of embedding & overpacking cementitious matrices
    - H_2 gas generation and release (source term) → facility safety
    - Gas overpressures (wp mechanical behaviour) → wp bursting
  - Concrete container air carbonation
    - Low pH propagation front → reinforcements depassivation = corrosion
    - Calcite ($CaCO_3$) precipitation → radiolytic H_2 gas release lowering
  - Degradation of reinforced concrete related to corrosion
    - Expansive corrosion products formation → wp damaging
    - Alteration of wp confinement property
    - Wp recovery impossibility
    - Waste-form re-encapsulation (disposal entrance phase compatibility)?

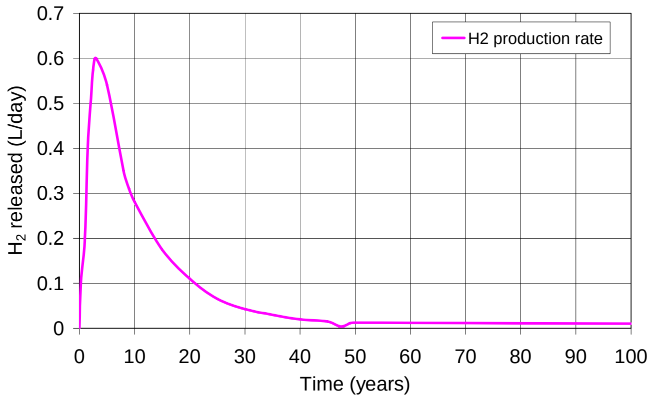
Radiolysis: decomposition of pore-water by ionizing radiations

- Estimation of the H_2 gas wp source term (chemo-transport coupling)
- Evaluation of the wp gas overpressurization risk (mechanical effect)

Data analysis of real waste package tests case + simulation
Validation experiments → model robustness evaluation (*CHEMSIMUL*)





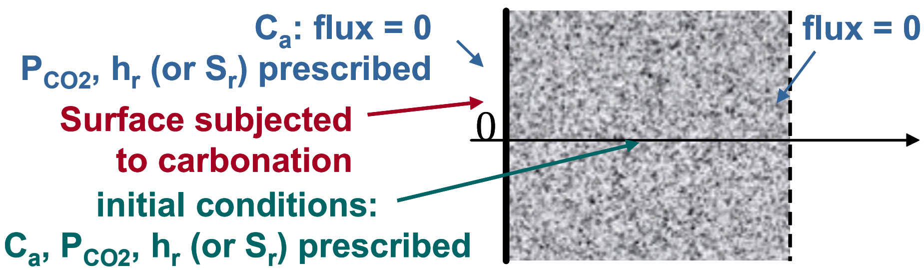


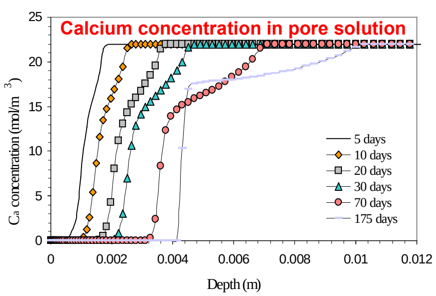
(Bouniol, 2004)

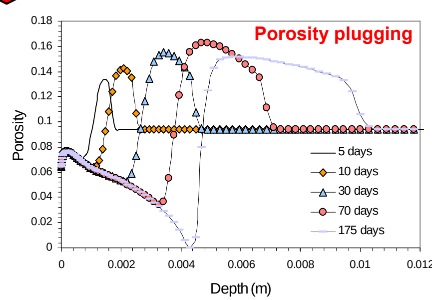
- **Air carbonation: a key factor for corrosion process of reinforced concrete structures (pH drop at the reinforcement/concrete interface)**
  - Evaluation of the natural carbonation front propagation kinetics
  - → Concrete properties, temperature and relative humidity conditions

Development of a simplified chemo-transport model

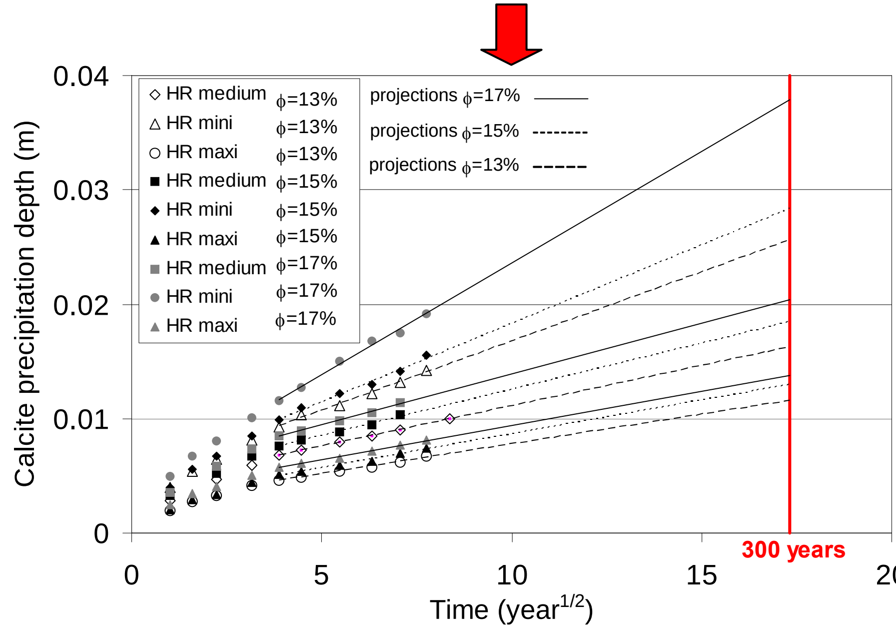
Experimental validation through accelerated carbonation tests and field data







Long-term prediction



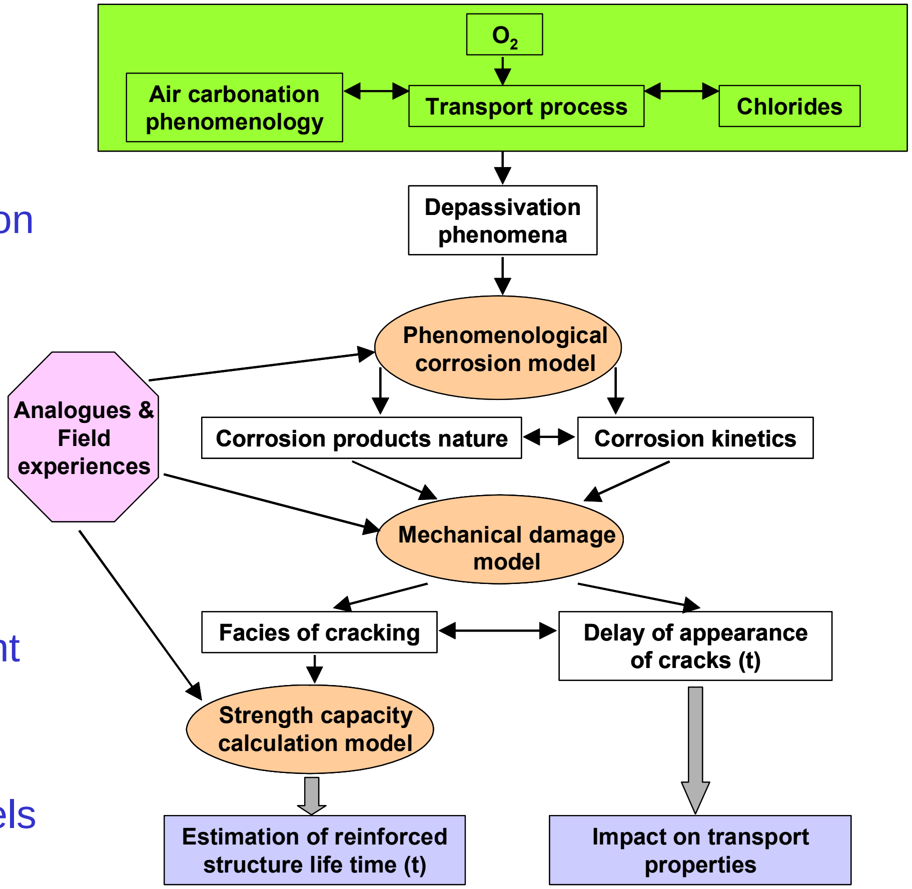
(Bary, 2005)

Corrosion of reinforced concrete structures: CIMETAL Project

- **Operational objectives:** concrete structures life-time evaluation & ruin prevention
- **Scientific objectives:** phenomenology understanding, models development & validation

Main scientific aspects:

- **CIM 1: Phenomenology**
  - Corrosion **rates** & **products** + transition conditions between passive - active states
  - **Corrosion model** development → unsaturated conditions (carbonation)
- **CIM 2: Mechanical behaviour**
  - **Mechanical impact** of corrosion products growth (data acquisition)
  - Concrete **damage model** development
- **CIM 3: Long-term prediction**
  - Phenomenological knowledge & models validation through **field experiences**

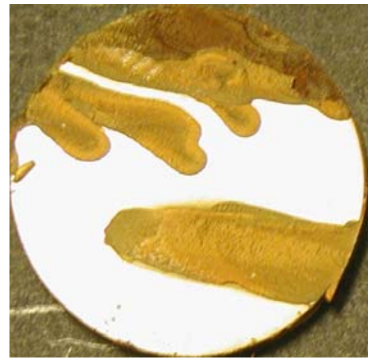


(L'Hostis et al., 2005)

- **Concrete pore-water / FeE500 corrosion rates and products**

$CaCO_3$, pH=8.3 V_{corr}=300 µm/y.
Lepidocrocite, magnetite

$CaCO_3 + SiO_2$ (am), pH=8.3 V_{corr}=180 µm/y.
Magnetite, siderite

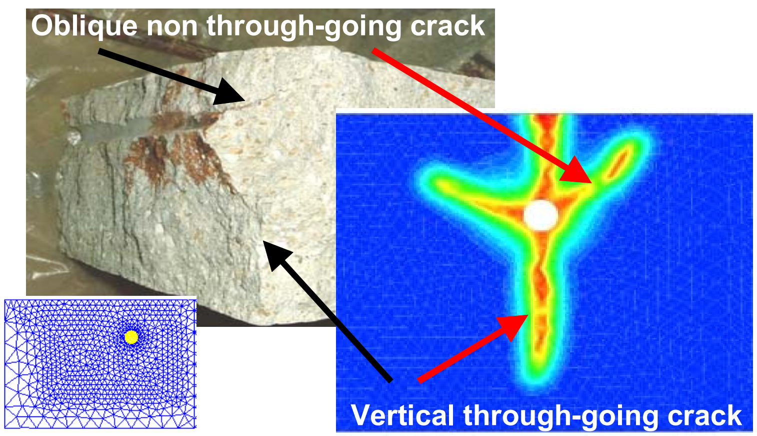


$CaCO_3 + SiO_2$ (am) + $NaHCO_3$, pH=9.1 V_{corr}=80 µm/y. (GR CO_3^{2-})

$CaCO_3 + SiO_2$ (am) + $NaHCO_3$ + $CaSO_4$, pH=8.0 V_{corr}=80 µm/y.
Calcite, Ferroxyhite?

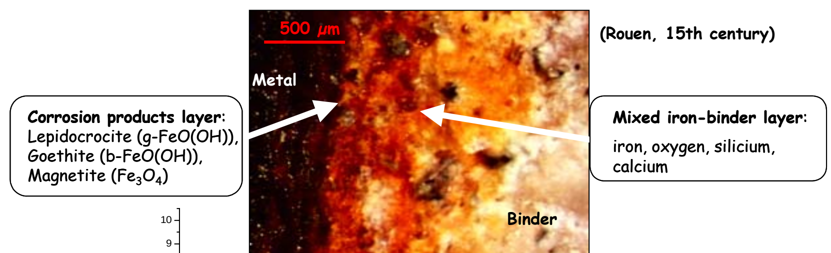
(Huet et al., 2004)

- **Damage modelling: through-going cracks appearance delay (exp. / sim.)**

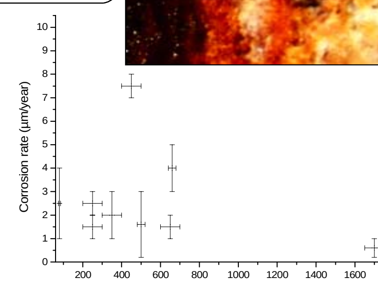


(Millard et al., 2004)

- **Estimation of average corrosion rates in natural analogue systems**



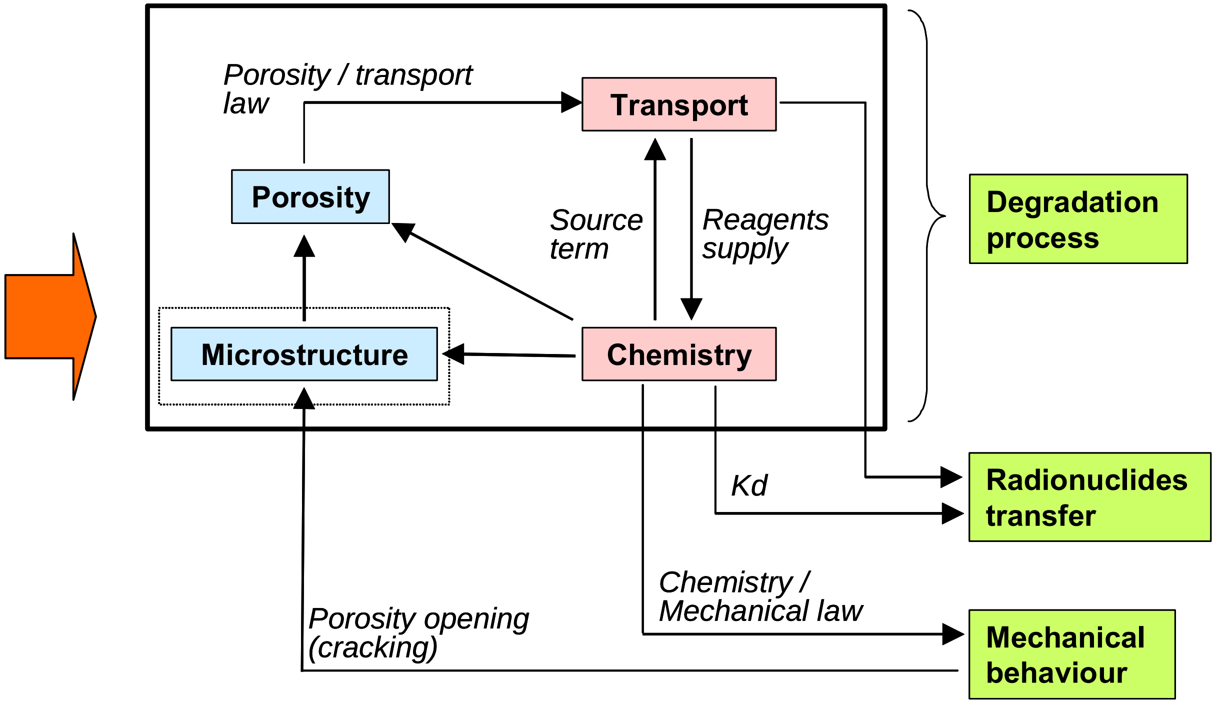
(Rouen, 15th century)



(Chitty et al., 2005)

- **Key identified topics for the *wp* long-term disposal evolution**
  - Concrete leaching by underground waters leading to:
    - Chemical degradation of cementitious phases
    - Dissolution / precipitation processes
    - Microstructure and transport properties evolution (feedback effect)
    - Mechanical effect (expansive phenomena, cracking…)
    - Impact on radionuclides (RN) transport

Chemical evolution, transport properties, mechanical performances and RN transport are strongly coupled problematics

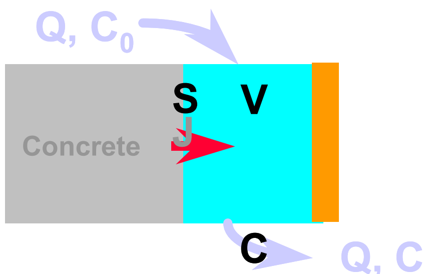


Cement-based materials leaching studies in pure water

- **Objective:**
  - Prediction of cement-based materials chemical degradation in relation with mineralogical, microstructural and transport coefficient changes

- **Scientific approach means:**
  - Phenomenological knowledge of key phenomena and associated parameters
  - Models development & numerical simulation (chemistry-transport coupling)

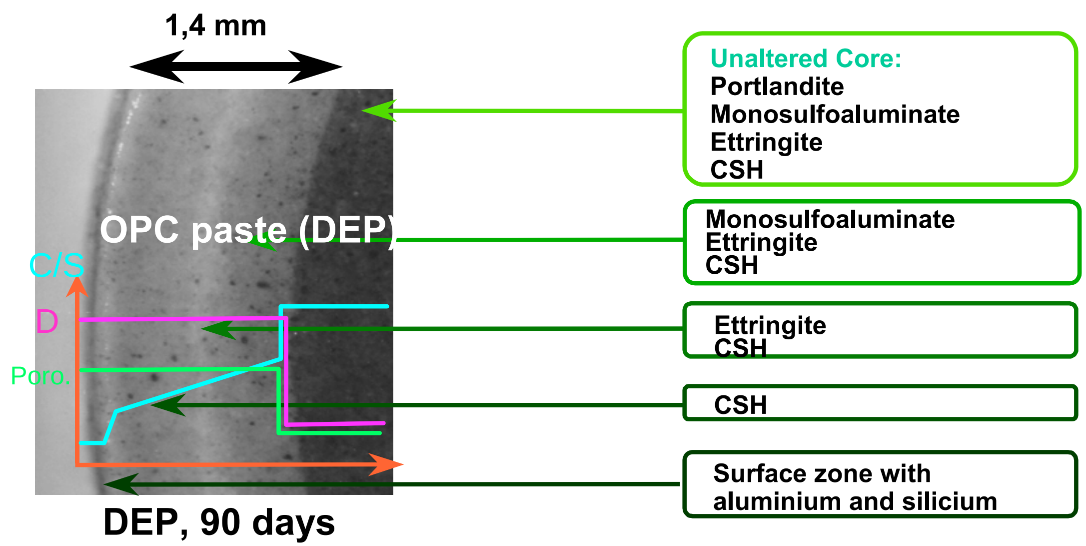
Ca^{2+} and OH^- are the main leached species / Migration is controlled by diffusion process



$$\frac{\partial \phi . C_{Ca}}{\partial t} = \frac{\partial (D . \phi . \partial C_{Ca})}{\partial (\partial x)} - \frac{\partial S_{Ca} (C_{Ca})}{\partial t}$$

$$V . \frac{dC}{dt} = S . J \Big|_{x=0} + Q . (C_0 - C)$$

$$J \Big|_{x=0} = - D \frac{\partial C}{\partial x} \Big|_{x=0}$$

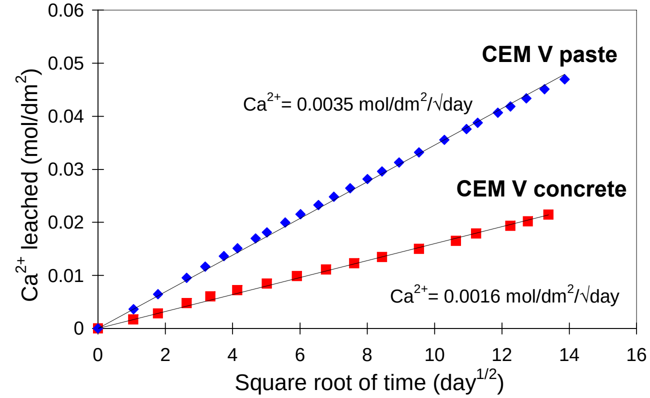


(Adenot, 1992; Le Bescop et al., 2000; Peycelon et al., 2001)

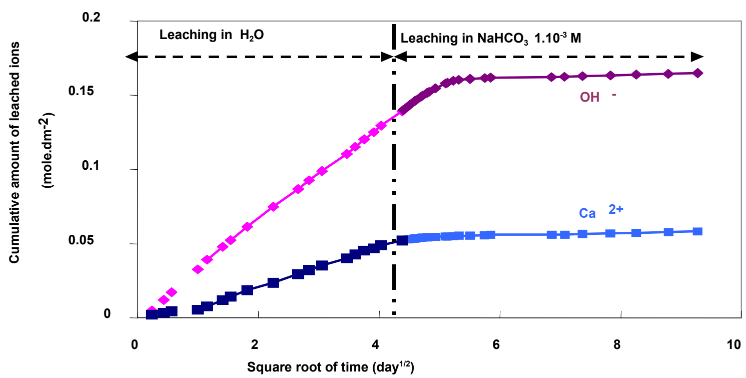
Leaching studies: materials and chemical environment influence

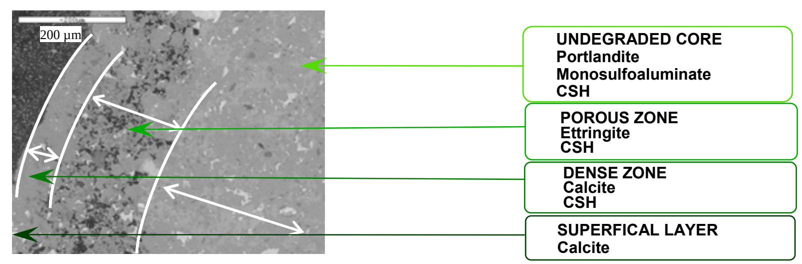
- **Leaching experiments & models validation**

T (°C)	Data type	CEM I / Ca^{2+} leached mol/dm^2/√day	CEM I degraded thickness mm/√day
25°C	Exp.	0.015	0.19
	Mod.	0.015	0.17
50°C	Exp.	0.025	0.29
	Mod.	0.026	0.31
80°C	Exp.	0.043	0.55
	Mod.	0.045	0.54



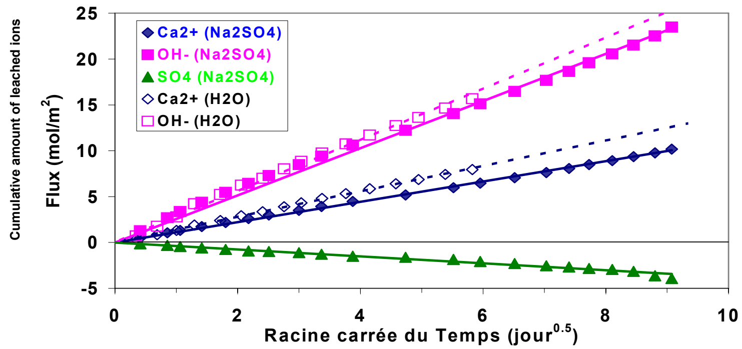
Carbonates

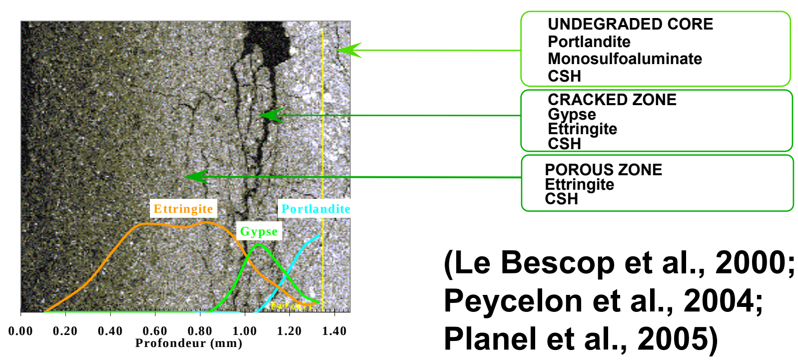




Pâte CEM I après 7 mois d 'altération

Sulphates



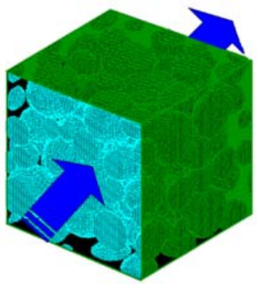


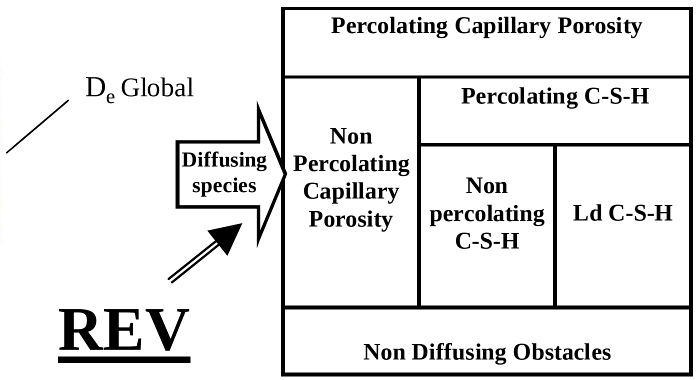
(Le Bescop et al., 2000; Peycelon et al., 2004; Planel et al., 2005)

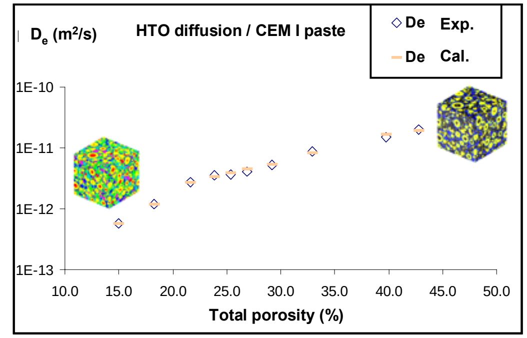
- **Microstructure – transport (diffusion) coupled approach (Microstrans)**
  - Hydration model + homogeneisation method: properties of an heterogeneous system (REV) based on elementary properties of components (microscale)

Phase 1 : Q_1, D_{ei1}

Phase 2 : Q_2, D_{ei2}

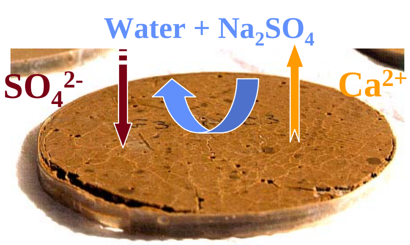


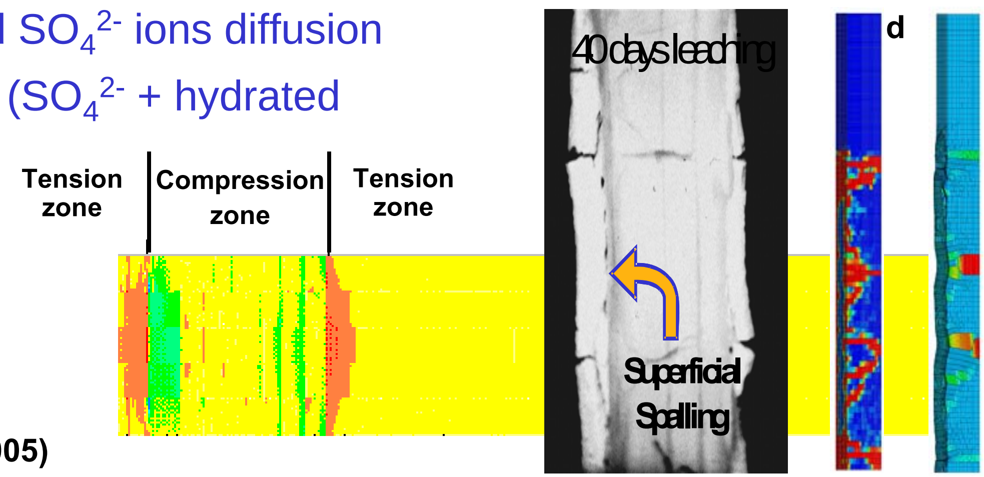




(Bejaoui et al., 2003)

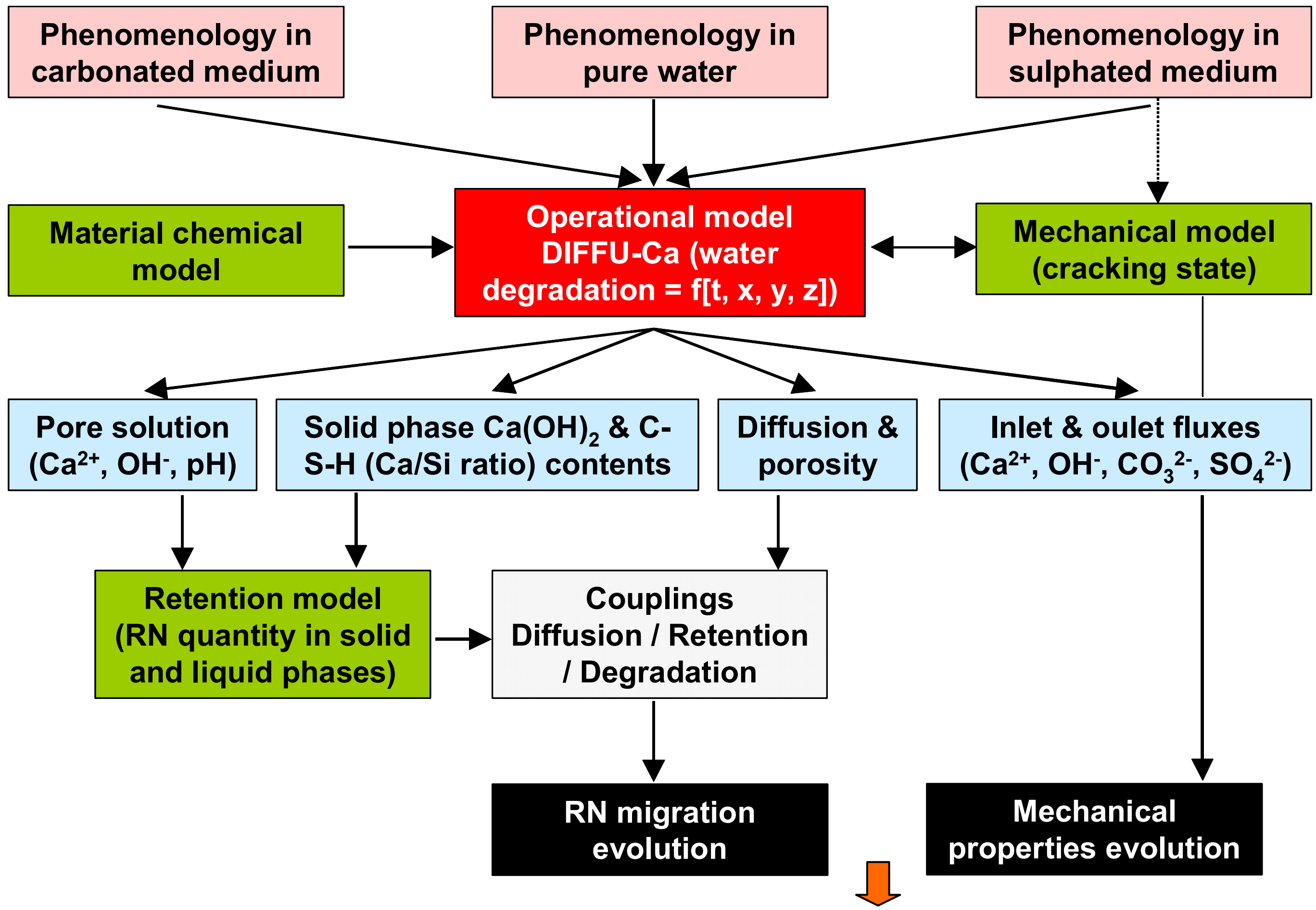
- **External sulphate attack: phenomenological understanding & CT-M modelling**
  - Cement paste decalcification and SO_4^{2-} ions diffusion
  - Gypsum & ettringite precipitation (SO_4^{2-} + hydrated and anhydrous aluminates)





(Planel et al., 2005)

Operatinal modelling approach (wp long-term behaviour)



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MOP 2005: decalcification + carbonation + cracking + RN transport + D_e=f(Ø)

(Peycelon et al., 2001; Richet et al., 2004)

- **Prediction of concrete structures and cemented waste package long-term evolution in storage and disposal context**

  - Scientific & operational strategy / approach:
    - Phenomenological understanding of dominant processes & mechanisms
    - Phenomenological models development
    - Phenomenologies coupling (ie. Hydrolysis, carbonates, sulphates, T°C…)
    - Chemistry, transport & mechanical processes coupling
    - Simplified modelling tools (MOP) to be integrated in numerical platform

  - Areas to be strengthened and associated main future works
    - Chemo-transport and mechanical coupling
    - Microstructure-diffusion model
    - Blended cement
    - Integration of “storage phase” = corrosion impact + (cracking)
    - MOP ability to describe real underground systems (to be strengthened)
    - Capacity to simulate the corrosion in degraded cement-based materials
    - Corrosion model coupling with a mechanical damage model